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(54) Polyarylene sulfide resin composition.

(57) A polyarylene sulfide composition is improved in impact resistance and comprises

(A) 100 parts by weight of a polyarylene sulfide resin and

(B) 0.5 to 50 parts by weight of a graft copolymer comprising (a) an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid and (b) one or more polymers or copolymers constituted of a repeating unit represented by the following general formula (1), both of which are chemically bonded to each other to form a branched or crosslinked structure:

$$-\!\left(CH_2 - \underset{\displaystyle X}{\overset{\displaystyle R}{\underset{|}{\overset{|}{C}}}} \right)\!- \qquad (1)$$

wherein R stands for a hydrogen atom or a lower alkyl group and X is an alkyl having 1 to 8 carbon atoms, straight or branched, phenyl or cyano.

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Description

POLYARYLENE SULFIDE RESIN COMPOSITION

The present invention relates to a polyarylene sulfide resin composition. Particularly, it relates to a polyarylene sulfide resin composition which is improved in impact resistance and is excellent in heat resistance and moldability.

[Prior Art]

Recently, a thermoplastic resin which has high heat resistance and chemical resistance and is excellent in flame retardance has been demanded as material for the components of electrical or electronic appliances, automobile devices or chemical instruments. A polyarylene sulfide resin represented by polyphenylene sulfide is one of the resins satisfying this demand and low costs for its physical properties, so that the demand therefor has increased.

However, a polyarylene sulfide resin has essential disadvantages in that it is poor in toughness and is brittle and that it is poor in mechanical properties represented by impact resistance.

Known methods for overcoming these disadvantages include:

(1) a method of adding a reinforcement such as glass fiber or carbon fiber,

and

(2) a method of adding other polymer.

However, these methods are not satisfactory enough owing to their inherent problems.

Namely, according to the method (1), although a polyarylene sulfide resin is remarkably improved in strength, stiffness, heat resistance and dimensional stability so as to permit the use thereof as an engineering plastic, the polyarylene sulfide resin composition according to the method (1) is inferior to other engineering plastics such as polyacetal, PBT, polysulfone or polyether sulfone in elongation, toughness and impact resistance, so that the use thereof is limited. On the other hand, although many attempts have been made with respect to the method (2), most of the attempts bring about lowering in the thermal deformation temperature and the results of the follow-up made by the inventors of the present invention revealed that few of them could exhibit a sufficient effect. Among them, for example, the addition of an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid as described in Japanese Patent Laid-Open Nos. 154757/1983, 152953/1984 and 189166/1984 is more effective to reduce the above disadvantages relatively efficiently. However, in many cases, the resulting composition exhibits insufficient impact resistance as yet, so that additional improvement is required.

[Summary of the Invention]

The inventors of the present invention have eagerly studied for the purpose of further enhancing the effect brought about by the above addition of an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid to thereby improve the impact resistance of a polyarylene sulfide resin more satisfactorily and have found that the impact resistance of a polyarylene sulfide resin can be remarkably improved not by the above-mentioned addition of a copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid alone, but by the addition of a graft copolymer obtained by chemically bonding a specified polymer to such a copolymer to form a branched or crosslinked chain. The present invention has been accomplished on the basis of this finding.

Namely, the present invention relates to a polyarylene sulfide resin composition comprising

(A) 100 parts by weight of a polyarylene sulfide resin,

(B) 0.5 to 50 parts by weight of a graft copolymer comprising (a) an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid and (b) one or more polymers or copolymers constituted of a repeating unit represented by the following general formula (1), both of which are chemically bonded to each other to form a branched or crosslinked structure:

$$-\!\left(CH_2 - \underset{\displaystyle X}{\underset{|}{\overset{\displaystyle R}{\overset{|}{C}}}} \right)\!- \qquad (1)$$

wherein R stands for a hydrogen atom or a lower alkyl group and X is an alkyl having 1 to 8 carbon atoms, straight or branched, phenyl or cyano, the alkyl preferably including

$-COOCH_3$, $-COOC_2H_5$, $-COOC_4H_9$, and

$-COOCH_2CH(C_2H_5)C_4H_9$,

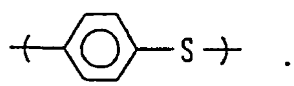
and

(C) 0 to 400 parts by weight of an inorganic filler.

The resin (A) to be used as the base in the present invention is a polyarylene sulfide resin comprising at least 70 molar % of a repeating unit represented by the structural formula -(Ar-S)- (wherein Ar is an aryl group) (hereinafter this resin will be abbreviated to "PAS"). A representative example thereof is polyphenylene sulfide comprising at least 70 molar % of a repeating unit represented by the structural formula: -(Ph-S)- wherein Ph is a phenyl group) (hereinafter this resin will be abbreviated to "PPS"). Particularly, it is suitable to use a polyphenylene sulfide having a melt viscosity as determined at 310°C with a shear rate of 1200/sec of 10 to 20000 P, preferably 100 to 5000 P.

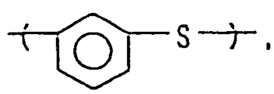
Known polyphenylene sulfide resins are classified into two groups. One group comprises substantially linear ones free from branched or crosslinked structure, while the other group comprises ones having a branched or crosslinked structure, the difference in the structure resulting mainly from the preparation process thereof. Although the present invention is effective for both the groups, it is more effective for linear ones free from any branched structure.

Preferred examples of the PPS polymer to be used in the present invention include polymers comprising at least 70 molar %, still preferably at least 80 molar %, of a p-phenylene sulfide unit:

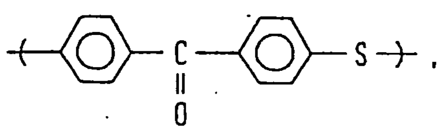


If the content of this unit is less than 70 molar %, the crystallinity of the resulting polymer will be too low to attain a sufficiently high strength and the toughness thereof will be also poor, though high crystallinity is a characteristic of a crystalline polymer.

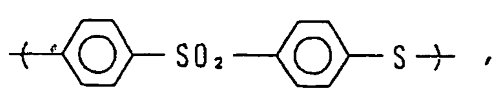
The linear PSS polymer to be used in the present invention may contain at most 30 molar % of another comonomer unit and examples of the comonomer unit include a m-phenylene sulfide unit:



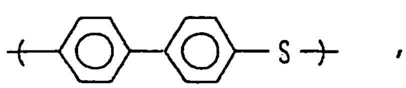
a diphenyl ketone sulfide unit:



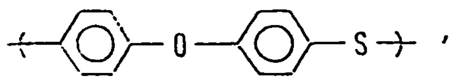
a diphenyl sulfone sulfide unit:



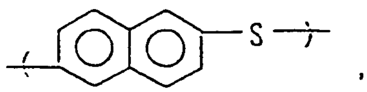
a diphenyl sulfide unit



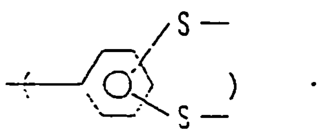
a diphenyl ether sulfide unit:



a 2,6-naphthalene sulfide unit:

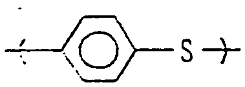


and a trifunctional unit:

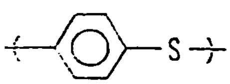


It is preferable that the content of a trifunctional unit be 1 molar % or below.

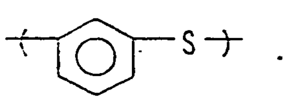
Particularly preferred examples of the PPS polymer to be used in the present invention include a linear PPS homopolymer composed of p-phenylene sulfide units:



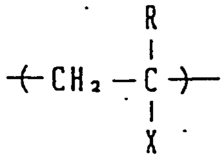
and linear block copolymers comprising 70 to 95 molar % of p-phenylene sulfide repeating units:



and 5 to 30 molar % of m-phenylene sulfide repeating units:

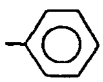


The graft copolymer (B) to be used in the present invention is a graft copolymer comprising (a) an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid and (b) one or more polymers or copolymers constituted of a repeating unit represented by the following general formula (1), both of which are chemically bonded to each other to form a branched or crosslinked structure:



(1)

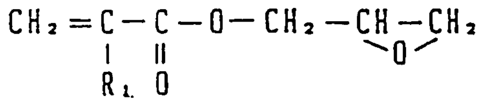
wherein R stands for a hydrogen atom or a lower alkyl group and X is an alkyl having 1 to 8 carbon atoms, straight or branched, such as
$-COOCH_3$, $-COOC_2H_5$, $-COOC_4H_9$, and $-COOCH_2CH(C_2H_5)C_4H_9$,



,

-CN.

The α-olefin which is one of the monomers constituting the copolymer (a) includes ethylene, propylene and butene-1, among which ethylene is preferably used. The glycidyl ester of an α,β-unsaturated acid which is the other of the monomers constituting the copolymer (a) is represented by the general formula (2):



(2)

wherein R_1 stands for a hydrogen atom or a lower alkyl group.
Examples thereof include glycidyl acrylate, glycidyl methacrylate and glycidyl ethacrylate, among which glycidyl methacrylate is preferably used. The copolymerization of an α-olefin (such as ethylene) with a glycidyl ester of an α,β-unsaturated acid may be carried out by a conventional radical polymerization method to obtain a copolymer (a).

It is preferable that the segment (a) comprise 70 to 99% by weight of an α-olefin and 30 to 1% by weight of a glycidyl ester of an unsaturated acid.

The polymer or copolymer (b) to be grafted onto the copolymer (a) to form a branched or crosslinked chain is a polymer comprising a repeating unit represented by the general formula (1) or a copolymer comprising two or more repeating units represented thereby, and examples thereof include polymethyl methacrylate, polyethyl acrylate, polybutyl acrylate, poly-2-ethylhexyl acrylate, polystyrene, polyacrylonitrile, polyacrylonitrile-styrene copolymers, polybutyl acrylate-polymethyl methacrylate copolymers and polybutyl acrylate-polystyrene copolymers. These polymers and copolymers can be each prepared by the radical polymerization of corresponding vinyl monomers.

The component (B) to be characteristically used in the present invention is neither the above copolymer (a) nor the (co)polymer (b), but a graft copolymer comprising the copolymer (a) and the (co)polymer, (b), both of which are chemically bonded to each other at at least one point to form a branched or crosslinked structure. In virtue of this grafted structure, the component (B) attains a remarkable effect which could not be attained by the copolymer (a) or the (co)polymer (b) alone.

Although the process for preparing the graft copolymer from the segments (a) and (b) is not particularly limited, the graft copolymer can be easily prepared by radical reaction. For example, a free radical is formed in the polymer (b) by the use of, e.g., a peroxide and the resulting polymer (b) is melt kneaded with the segment (a) to obtain a graft copolymer (B). It is suitable that the graft copolymer (B) comprises the segments (a) and (b) in a ratio of 95 : 5 to 40 : 60.

The amount of the component (B) per 100 parts by weight of the component (A) is 0.5 to 50 parts by weight, preferably 1 to 20 parts by weight.

If the amount of the component (B) is too small, the objective improvement in the toughness and impact resistance will be hardly attained, while if the amount is too large, the thermal deformation temperature will be low and the mechanical properties such as stiffness will be unfavorably affected adversely.

Although the inorganic filler (C) is not necessarily an essential component in the present invention, the addition thereof is preferable for producing a molded article which is excellent in mechanical strength heat resistance, dimensional stability (resistance to deformation and warp), electrical properties and other performances. The inorganic filler (C) may be selected from among fibrous, powdery, granular and flaky ones

depending upon the object.

Examples of suitable fibrous fillers include inorganic fibrous materials, for example, glass fiber, asbestos fiber, carbon fiber, silica fiber, silica/alumina fiber, zirconia fiber, boron nitride fiber, silicon nitride fiber, boron fiber, potassium titanate fiber and fibers of metals such as stainless steel, aluminum, titanium, copper or brass. Among them, glass fiber and carbon fiber are most representative. Further, the fibrous filler includes high-melting organic fibrous materials, and particular examples thereof include polyamides, fluororesins and acrylic resins.

The powdery or granular filler includes carbon black, silica, quartz powder, glass bead, glass powder, silicates such as calcium silicate, aluminum silicate, kaolin, talc, clay, diatomaceous earth and wollastonite; metal oxides such as iron oxides, titanium oxide, zinc oxide and alumina; metal carbonates such as calcium carbonate and magnesium carbonate; metal sulfates such as calcium sulfate and barium sulfate; silicon carbide, silicon nitride, boron nitride and various metal powders.

The flaky filler includes mica, glass flake and various metal foils.

These inorganic fillers may be used alone or as a mixture of two or more of them. The simultaneous use of a fibrous filler, particularly glass or carbon fiber, with a granular and/or flaky filler is particularly effective in producing an article which is excellent not only in mechanical strengths but also in dimensional accuracy and electrical properties.

If necessary, a coupling agent or surface treatment is preferably used together with these fillers. Examples thereof include functional compounds such as epoxy, isocyanate, silane and titanate compounds. These compounds may be applied to the filler prior to the preparation of the composition to carry out the surface treatment or coupling of the filler or may be added in the course of preparation thereof.

The amount of the inorganic filler is 0 to 400 parts by weight, preferably 10 to 300 parts by weight, per 100 parts by weight of the polyarylene sulfide resin (A). If the amount is less than 10 parts by weight, the mechanical strengths will be slightly poor, while if it is too large, the molding will be difficult and the mechanical strengths of the resulting molded article will be problematic.

The composition of the present invention may auxiliarily contain a small amount of other thermoplastic resin in addition to the above components depending upon the object. This thermoplastic resin may be any one as far as it is stable at high temperature. Examples thereof include aromatic polyesters comprising aromatic dicarboxylic acids and diols or hydroxy carboxylic acids, such as polyethylene terephthalate and polybutylene terephthalate; polyamide, polycarbonate, ABS, polyphenylene oxide, polyalkyl acrylate, polyacetal, polysulfone, polyether sulfone, polyether imide, polyether ketone and fluororesins. These thermoplastic resins may be used also as a mixture of two or more of them.

Further, the composition of the present invention may suitably contain an additive which is conventionally added to a thermoplastic or thermosetting resin, depending upon the performance required. Examples of such an additive include stabilizers such as antioxidant and ultraviolet absorber, antistatic agent, flame retardant, coloring agent such as dye and pigment, lubricant, crystallization accelerator and nucleating agent.

The polyarylene sulfide resin composition of the present invention can be prepared by a conventional process for the preparation of a synthetic resin composition and with conventional equipment therefor. Namely, necessary components are mixed and kneaded and extruded with a single- or twin-screw extruder to obtain a pellet. In the preparation, a part of the necessary components may be mixed, as a master batch, with the residual part thereof, followed by molding. Alternatively, a part or the whole of the resinous components may be preliminarily ground, followed by mixing and extrusion.

As described in the above description and Examples, the polyarylene sulfide resin composition of the present invention is remarkably improved in mechanical properties, particularly toughness and impact resistance as compared with the one of the prior art and exhibits reduced lowering in the thermal deformation temperature, though compositions similar to the one of the present invention generally exhibit significant lowering therein. Thus, the composition of the present invention is expectable as a material suitable for various functional components.

[Example]

The present invention will now be described in more detail by referring to the following Examples, though it is not limited by them.

Examples 1 to 11 and Comparative Examples 1 to 7

Each of the compounds given in Tables 1 to 3 of an amount given therein was added, as the component (B), to a PSS resin ("Fortlon KPS"; a product of Kureha Chemical Industry Co., Ltd.) as the component (A), followed by premixing with a Henschel mixer for 5 minutes. A commercially available glass fiber (diameter: 13 µm, length: 3 mm) and/or calcium carbonate (average particle size: 4 µm) were added each in an amount given in Tables 1 to 3 to the obtained premix, followed by mixing for 2 minutes. The obtained mixture was extruded with an extruder at a cylinder temperature of 310°C to obtain pellets of a polyphenylene sulfide resin composition. The pellets were molded into an ASTM test piece with an injection molding machine at a cylinder temperature of 320°C and a mold temperature of 150°C. This piece was examined for physical properties. The results are shown in Tables 1 to 3.

As Comparative Examples, in a similar manner to the one described above, the preparation of a composition was carried out except that no component (B) was added or the component (a) or (b) was added alone as the

component (B), and the obtained composition was molded into a test piece and examined. The results are shown in Tables 1 to 3.

The evaluation items and the method of measurement thereof are as follows:

Izod impact strength: determined according to ASTM D-256 with respect to both notched and unnotched sides.

Thermal deformation temperature: determined according to ASTM D-648 (under a load of 18.6 kg/cm^2).

Tensile test: Tensile strength and tensile elongation were determined according to ASTM D-638.

Table 1

Composition		Ex. 1	Ex. 2	Comp. Ex. 1	Comp. Ex. 2
(A)	PPS (parts by weight)	95	90	100	90
(C)	glass fiber (diameter: 13 μm, length: 3 mm) (parts by weight)	-	-	-	-
	calcium carbonate (average particle size: 4 μm) (parts by weight)	-	-	-	-
(B)	E/GMA*1-polymethyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polymethyl methacrylate/butyl acrylate (70-30) graft copolymer (parts by weight)	5	10	-	-
	E/GMA*1-polystyrene (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polyacrylonitrile/styrene (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polybutyl acrylate/styrene/methyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polyethyl acrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
(a)	E/GMA*1 (alone) (parts by weight)	-	-	-	10
Physical properties	Izod impact strength, notched side (kg·cm/cm)	3.7	5.2	2.5	3.7
	Izod impact strength, unnotched side (kg·cm/cm)	17.0	21.1	12.0	16.8
	theraml deformation temperature (°C)	-	-	-	-
	tensile strength (kg/cm^2)	833	745	898	700
	tensile elongation (%)	5.7	6.9	4.0	6.1

*1 E/GMA: ethylene/glycidyl methacrylate (85:15) copolymer

Table 2

Composition		Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Comp. Ex. 3	Comp. Ex. 4	Comp. Ex. 5
(A)	PPS (parts by weight)	55	50	50	50	50	50	50	60	50	50
(C)	glass fiber (diameter: 13 µm, length: 3 mm) (parts by weight)	40	40	40	40	40	40	40	40	40	40
	calcium carbonate (average particle size: 4 µm) (parts by weight)	-	-	-	-	-	-	-	-	-	-
(B)	E/GMA*1-polymethyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	10	-	-	-	-	-	-	-
	E/GMA*1-polymethyl methacrylate/butyl acrylate (70-30) graft copolymer (parts by weight)	5	10	-	-	-	-	-	-	-	-
	E/GMA*1-polystyrene (70-30) graft copolymer (parts by weight)	-	-	-	10	-	-	-	-	-	-
	E/GMA*1-polyacrylonitrile/styrene (70-30) graft copolymer (parts by weight)	-	-	-	-	10	-	-	-	-	-
	E/GMA*1-polybutyl acrylate/styrene/methyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	-	-	-	10	-	-	-	-
	E/GMA*1-polyethyl acrylate (70-30) graft copolymer (parts by weight)	-	-	-	-	-	-	10	-	-	-
(a)	E/GMA*1 (alone) (parts by weight)	-	-	-	-	-	-	-	-	10	-
(b)	polymethyl methacrylate/butyl acrylate copolymer (alone) (parts by weight)	-	-	-	-	-	-	-	-	-	10
Physical properties	Izod impact strength, notched side (kg·cm/cm)	13.5	16.5	14.8	16.1	15.5	16.6	13.3	8.5	12.2	11.1
	Izod impact strength, unnotched side (kg·cm/cm)	54.5	58.2	50.5	56.8	51.9	57.3	55.2	37.1	45.8	42.5
	thermal deformation temperature (°C)	267	266	265	265	265	266	267	269	260	258
	tensile strength (kg/cm^2)	1640	1460	1480	1520	1510	1440	1490	1680	1320	1250
	tensile elongation (%)	2.0	2.3	2.0	2.2	2.1	2.2	2.0	1.6	1.9	1.8

*1 E/GMA: ethylene/glycidyl methacrylate (85:15) copolymer

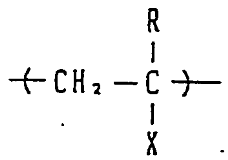
Table 3

Composition		Ex. 10	Ex. 11	Comp. Ex. 6	Comp. Ex. 7
(A)	PPS (parts by weight)	29	24	34	24
(C)	glass fiber (diameter: 13 μm, length: 3 mm) (parts by weight)	33	33	33	33
	calcium carbonate (average particle size: 4 μm) (parts by weight)	33	33	33	33
(B)	E/GMA*1-polymethyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polymethyl methacrylate/butyl acrylate (70-30) graft copolymer (parts by weight)	5	10	-	-
	E/GMA*1-polystyrene (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polyacrylonitrile/styrene (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polybutyl acrylate/styrene/methyl methacrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
	E/GMA*1-polyethyl acrylate (70-30) graft copolymer (parts by weight)	-	-	-	-
(a)	E/GMA*1 (alone) (parts by weight)	-	-	-	10
Physical properties	Izod impact strength, notched side (kg·cm/cm)	7.8	10.3	5.8	7.6
	Izod impact strength, unnotched side (kg·cm/cm)	21.1	24.6	15.6	18.8
	thermal deformation temperature (°C)	270	270	271	268
	tensile strength (kg/cm^2)	1270	1190	1346	1090
	tensile elongation (%)	1.0	1.3	0.7	0.9

*1 E/GMA: ethylene/glycidyl methacrylate (85:15) copolymer

Claims

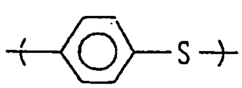
1. A polyarylene sulfide resin composition comprising
(A) 100 parts by weight of a polyarylene sulfide resin,
(B) 0.5 to 50 parts by weight of a graft copolymer comprising (a) an olefin copolymer comprising an α-olefin and a glycidyl ester of an α,β-unsaturated acid and (b) one or more polymers or copolymers constituted of a repeating unit represented by the following general formula (1), both of which are chemically bonded to each other to form a branched or crosslinked structure:



(1)

wherein R stands for a hydrogen atom or a lower alkyl group and X stands for one or more members selected from among an alkyl having 1 to 8 carbon atoms, straight or branched, phenyl and cyano, and
(C) 0 to 400 parts by weight of an inorganic filler.

2. A polyarylene sulfide resin composition as set forth in claim 1, wherein said polyarylene sulfide resin is a poly-p-phenylene sulfide containing at least 70% by weight of a repeating unit represented by the formula:



3. A polyarylene sulfide resin composition as set forth in claim 2, wherein the said poly-p-phenylene sulfide contains at least 80% by weight of the said repeating unit.

4. A polyarylene sulfide resin composition as set forth in any preceding claim, wherein the polyarylene sulfide resin (A) is selected from a homopolymer composed of p-phenylene sulfide units and a linear block copolymer comprising 70 to 95 molar % of p-phenylene sulfide units and 5 to 30 molar % of m-phenylene sulfide units.

5. A polyarylene sulfide resin composition as set forth in any preceding claim, wherein said olefin copolymer (a) constituting the component (B) is a copolymer comprising ethylene and a glycidyl ester of an α,β-unsaturated acid.

6. A polyarylene sulfide as set forth in any preceding claim, wherein the segment (a) of the component (B) comprises 70 to 99% by weight of the α-olefin and 30 to 1 % by weight of the glycidyl ester.

7. A polyarylene sulfide resin as set forth in any preceding claim, wherein the amount of the component (B) per 100 parts by weight of the component (A) is 1 to 20 parts by weight.

8. A polyarylene sulfide resin composition as set forth in claim 1, 2 or 3, wherein the amount of the inorganic filler (C) added is 10 to 300 parts by weight.

9. A polyarylene sulfide resin composition as set forth in claim 1, 2, 3 or 4, wherein said inorganic filler is a fibrous one.

10. A polyarylene sulfide resin composition as set forth in claim 1, 2, 3, 4 or 5, wherein said inorganic filler (C) is a combination of a fibrous one with a powdery or granular one.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 30 0906

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	EP-A-0 247 412 (BAYER AG) * claims * ---	1-10	C 08 L 81/02 C 08 L 51/06
A	EP-A-0 228 268 (TORAY) * claims * ---	1-10	
A	PATENT ABSTRACTS OF JAPAN vol. 12, no. 15 (C-469)(2862), 16th January 1988; & JP - A - 62 172 057 (TORAY) 29-07-1987 -----	1	

TECHNICAL FIELDS SEARCHED (Int. Cl.4)
C 08 L 81/00 C 08 L 51/00

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	26-04-1989	BOEKER R.B.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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